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### SNM Detection with a Large Water Cerenkov Detector

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#### Abstract:

Special Nuclear Material (SNM) can either spontaneously fission, or be induced to do so. Either case results in neutron emission. Since neutrons are highly penetrating and difficult to shield, they could, potentially, be detected escaping even a well shielded cargo container. Obviously, if the shielding is sophisticated, detecting it would require a highly efficient detector with close to  $4\pi$  solid angle coverage. Water Cerenkov detectors may be a cost effective way to achieve that goal if it can be shown that the neutron capture signature is large enough and if sufficient background rejection can be employed as detectors get larger. In 2008 the LLNL Advanced Detector Group reported the successful detection of neutrons with a ¼ ton gadolinium doped water Cerenkov prototype. We have now built a 4 ton version. This detector is not only bigger, it was designed with photon detection efficiency in mind from the beginning. We are employing increased photocathode coverage and more reflective walls, coated with PTFE. The increased efficiency should allow better energy resolution. We expect that the better diffusive wall reflectivity will reduce the overall dependence of the detector response on particle direction, again producing a more consistent response. We also believe that as detectors get larger, both uncorrelated and correlated backgrounds due to gamma-rays and cosmic ray interactions near the detector will increase. To prove the effectiveness of the technology we must develop new ways to reject these backgrounds while maintaining our sensitivity to SNM neutrons. Better energy resolution will enable us to reject more of the low energy gamma-ray backgrounds on this basis. Overcoming cosmic ray induced neutrons is perhaps an even larger concern as detectors get larger. Our detector is designed so that we can test various segmentation schemes – effectively dividing the detector up into smaller ones. In this presentation, we will describe our detector in detail.

**Keywords:** Water Cerenkov, neutron detector, gadolinium, neutron capture

#### **Introduction:**

Legitimate cross border trade involves the transport of an enormous number of cargo containers. Especially following the September 11 attacks, verifying that these containers are not transporting Special Nuclear Material (SNM) has become a priority of both national and international security. This must be done without impeding legitimate trade. To meet both needs, a large number of fast, highly efficient, easy to operate and relatively inexpensive detectors must be deployed at border crossings and ports. The U.S. and other countries have already begun such deployments. However, there remains a need for improved detectors which can maintain sensitivity to SNM while reducing false positive

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or negative detections, and without undue cost or complexity. A related need is to limit sensitivity to other types of background radiation, such as cosmic-ray induced background or any Naturally Occurring Radioactive Material (NORM) that may be present in certain legitimate forms of cargo. The detector development work described here is intended to provide such improvements for commercial screening (and possibly other) nuclear security applications.

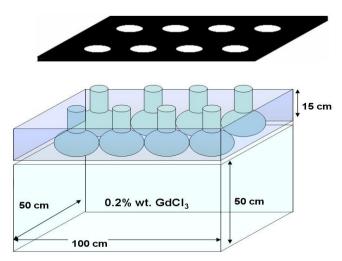
Because the signature of SNM, crudely speaking, consists of several neutrons and gamma rays emitted isotropically and in close time proximity, large solid angle coverage is imperative. This recognition has led to the use of large organic plastic or liquid scintillator detectors [1 - 4]. These devices have a relatively poor spectral response. As a result, these methods often rely on detecting an increase in the rate of incoming particles compared to background, rather than the analysis of the particle energy spectrum. Even without spectroscopy, rate-based approaches, such as the 'nuclear car wash' interrogation scheme [4], have been shown to be effective.

Although organic scintillators are attractive in many respects, they have several drawbacks. Plastic organic scintillators are expensive to build in large volumes, and difficult to dope with neutron capturing agents like Gd or <sup>6</sup>Li. Liquid scintillator is often toxic and highly flammable. Given these limitations water Cerenkov based detectors offer an interesting alternative. Water is non-toxic, non-flammable and inexpensive, and retains these properties even after doping with Gadolinium or other neutron absorbing compounds. For example, the Super-K and SNO experiments [5,6] have shown that the Cerenkov process can generate enough photons to permit detection of gamma-rays with an energy greater than about 3 to 4 MeV - or neutron captures on chlorine - so long as the photocathode coverage is high (~40%). Neutron capture on chlorine (like gadolinium) produces an 8 MeV gamma-ray cascade. SNO observed that the light output was approximately equivalent to a 6 MeV electron. As demonstrated here, the addition of highly reflective white walls makes it possible to detect gammas of a few MeV or more with a much smaller photocathode coverage - only about 10%.

Fission events from SNM produce a number of neutrons and MeV-scale gammas which are correlated over a wide range of times, from the few nanosecond level (prompt fission gammas) to the few hundred microsecond level (neutrons which thermalize in surrounding materials and capture in the detector). The observation of consistent time correlations between neutrons and gammas emitted from a cargo container could, therefore, constitute a robust signature for SNM, since this time coincident signature, although occurring at a low rate, stands out strongly against the uncorrelated gamma-ray backgrounds. Cosmogenic showers can also produce time-coincident signatures, but may be rejected since the showers are often accompanied by a high energy muon, which is easy to tag. One way to maximize the detection efficiency of these correlations would be to deploy a very large detector capable of near  $4\pi$  coverage around the cargo container – a "car wash" style detector. While "car wash" detectors are currently in development, most of the early concepts have relied on detecting either the fission gammas or regular decay gammas. Neutron detection remains more difficult, however, due to the high and potentially prohibitive expense of building many large "car wash" style <sup>3</sup>He detectors or

neutron sensitive liquid or plastic scintillator detectors. The Advanced Detectors Group at LLNL is investigating the use of gadolinium doped water as an alternative neutron detector medium, capable of realizing the construction of very large neutron and high energy gamma detectors, possibly at much lower cost than the alternatives. A water Cerenkov based detector would be relatively inexpensive if large size is required, and it should be possible to build a detector large enough to completely surround a cargo container. The detector might be useful in active or passive scenarios.

In addition to the size advantage that water based detectors may have, they are also relatively insensitive to low energy gamma-rays from NORM or the local environment, which depending on their energy, rarely or never create electrons above the Cerenkov threshold in water (0.78 MeV). Fast neutrons, which may be produced by nearby cosmic ray interactions, will produce a maximum of one capture signal per neutron. In a scintillator detector, a fast cosmogenic neutron has the potential to generate a correlated signal if a prompt/delayed pair is produced by nuclear recoil in the scintillator and a capture. The time between the two events will have a capture time characteristic to the scintillator, mimicking what may look like a correlated signal from SNM, arising from two coincident neutrons. This background is not a problem in a water Cerenkov detector, since fast-neutron-induced proton recoils within these detectors do not produce a prompt signal – except those caused by extremely rare multi GeV scale neutrons.

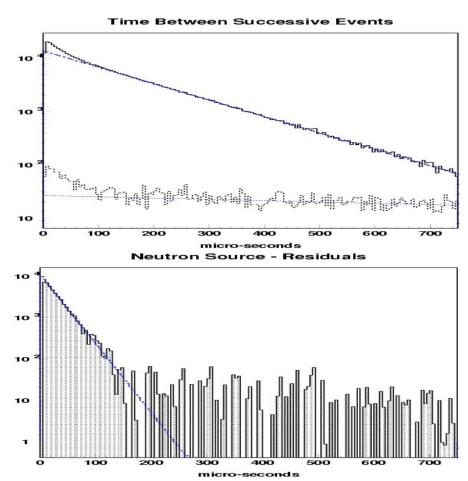


**Figure 1:** Schematic design of water Cerenkov detector (see text for description)

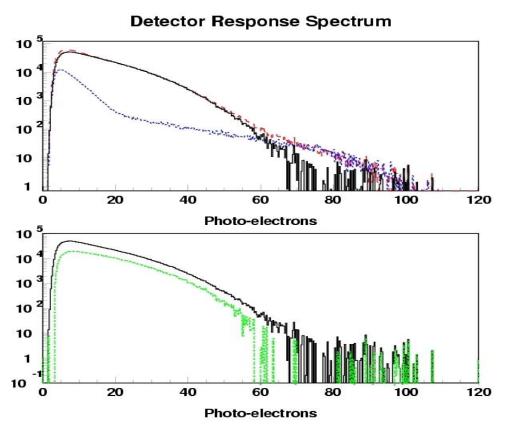
#### 250 Liter Prototype Detector

We recently established the feasibility of large water Cerenkov based neutron detectors with a 250 liter prototype detector (shown in Fig. 1) [1]. That detector used an ultra pure water detection medium doped with a small amount of gadolinium tri-chloride (0.2%), providing an overall gadolinium concentration of 0.1% by weight. Neutrons entering the detection medium were thermalized by the water and then captured on a gadolinium

nucleus. Gadolinium has a very large thermal neutron capture cross section (49000 barns). The thermalization process takes only a few microseconds. The capture time after thermalization follows an exponential distribution and in this case the average capture time was 28 microseconds.



**Figure 2:** The inter-event time distribution in the 250 liter prototype. The upper panel shows the resultant distributions with (upper histogram) and without (lower histogram) the presence of a <sup>252</sup>Cf source. The source results in an increase in both the random trigger rate and the correlated trigger rate. The lower panel shows the residuals resulting from the subtraction of random events, represented by an exponential fit at large inter-event times, leaving only the correlated component with a mean inter-event time of 28us.



**Figure 3:** The upper panel shows the summed PMT detector response spectra of our prototype with (dashed red line) and without (dotted blue line) the presence of a <sup>252</sup>Cf source. The difference between the two, representing the spectrum of a pure sample of <sup>252</sup>Cf source events (gammas and neutrons) in our prototype is shown as a solid black line in both the top and bottom panels. The green dot-dash line in the lower panel represents the statistically subtracted spectrum of pure neutron capture events in our detector.

Fig. 2 shows the inter-event time distribution between successive events with and without the presence of a  $^{252}$ Cf neutron source. The 55  $\mu$ Ci source was placed at a distance of one meter from the detector behind a two inch thick wall of lead. The presence of the source is easily discernable from the background in both the uncorrelated count rate – which increased from 700Hz to 7000Hz (Fig. 2a), and the correlated count rate, shown in Fig.2b.

Fig. 3 shows the spectral shape (in terms of photo-electrons detected) that results from the presence of neutron capture events in the detector.

## **A Four Ton Detector**

We have built a larger detector to test the efficiency of neutron detection as a function of size. This detector is approximately four tons in weight and represents an intermediate step between the 250 liter prototype and a full scale car wash style detector. It is important to show that the technology can be scaled up to any size without being swamped by unwanted backgrounds. A schematic of the detector is shown in Fig. 4 and some pictures of the construction phase are shown in Fig. 5. In a recent paper [8] we showed that water doped with 0.2% gadolinium tri-chloride seems to leach iron in small

quantities from stainless steel, severely limiting the transmission of near UV light in water. Therefore, all of the detector components were constructed from four plastics; polypropylene, polyethylene, acrylic and Teflon. The tank itself is a regular polyethylene water tank. PMT holders and a frame to support the PMTs on the top and bottom of the tank were constructed from acrylic and polypropylene. Since the whole PMT support structure floats, we used a ballast of stainless steel bars protected from the gadiated water by a two independently sealed polypropylene bags. The PMT cathode coverage has been increased to 19% from the 10% used in the prototype detector. The inside walls of the

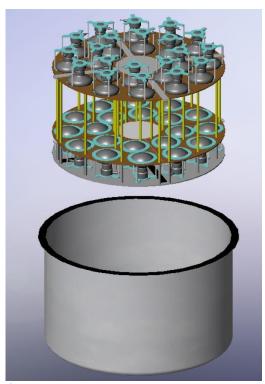
detector were surrounded by a layer of diffusively reflective Teflon to maximize photon reflections and hence detection efficiency.

# B. Backgrounds

Despite the successful detection of neutrons by the 250 liter prototype it is essential to show that the technology can achieve similar efficiencies as detectors get larger. Two problems that can potentially arise are the increased muon rate and subsequent dead time (if we veto such events) and increased correlated event rate due to cosmogenically generated neutrons. With this in mind the four ton detector has been designed allow segmentation into two or four optically separate zones. If, in fact, the correlated background rate is too high to run the detector monolithically, we intend to operate the detector in independent one or two ton segments.

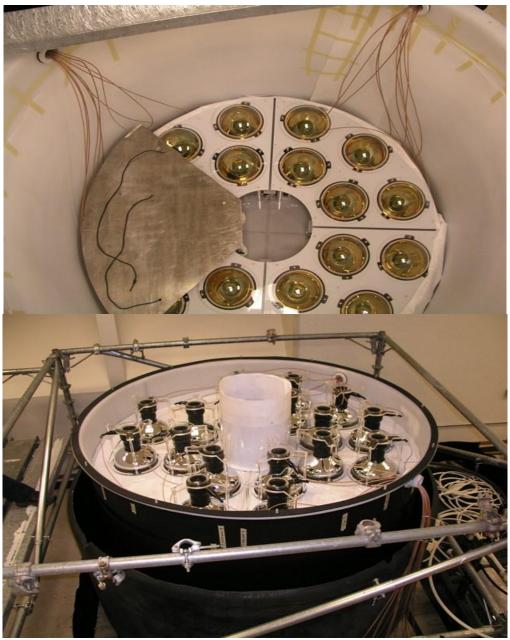
# C. Photon Collection efficiency

Another avenue for improvement is the photon collection efficiency. It is well known that Cerenkov light production is much less efficient than that of a liquid scintillator, generating only approximately 100 optical photons per MeV of electron energy. Additionally, what few photons are generated are channelled within a cone along the direction of the electron. Both the low number of photons and their directional nature reduce the energy resolution significantly. We propose to test a number of water soluble wavelength shifting chemicals to increase our resolution. The idea is to shift photons in the UV below about 300nm into the blue/green part of the spectrum, whilst simultaneously randomizing their direction. The overall effect will be to increase the number of photons available for collection by the PMTs and to smooth out non isotropic light. Tests carried out at SNO [9] indicate that some



**Fig. 4.** A schematic drawing of the four ton water Cerenkov detector. The detector has 40 ten inch PMTs arranged symmetrically on the top and bottom of the detector and supported by an acrylic/polypropylene frame. The frame is weighed down by approximately 400 lbs of stainless steel ballast protected from the water by a double layer of polypropylene bags.

wavelength shifting chemicals can potentially increase the number of collected photons by a factor of two to three.



**Figure 5:** The assembly of the PMT supporting cage within the new four ton detector. Top – shows the assembly of the internal frame that supports the PMTs. Middle – The bottom frame with PMTs installed. Note the aluminum step stool that fits around the PMT frame and enables access to the detector. Note also the spacers left for optical separators between each of the four quadrants. Bottom – The finished detector just prior to closing the top.

# **Conclusion:**

We have assembled two water Cerenkov neutron detectors. A small 250 liter detector filled with pure water doped with  $GdCl_3$  has been shown to detect neutrons emitted from a  $^{252}Cf$  source. On the strength of this result we undertook and completed the

construction of a new four ton detector of the same type. The detector will shortly undergo filling and testing. We will be testing the neutron detection efficiency at a number of positions located inside and outside the detector.

# **Acknowledgements:**

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#### **References:**

- [1] D. R. Slaughter, M. R. Accatino, A. Bernstein, et al., Nuclear Instruments and Methods in Physics Research B 241, 777, 2005
- [2] C. E. Moss, M. W. Brener, C. L. Hollas, W. L. Myers, Nuclear Instruments and Methods in Physics Research B 241, 793, 2005
- [3] J. Ely, R. Kouzes, J. Schweppe, et al., Nuclear Instruments and Methods in Physics Research A 560, 373, 2006
- [4] D. R. Slaughter, M. R. Accatino, A. Bernstein, et al., Nuclear Instruments and Methods in Physics Research A 579, 349, 2007
- [5] The Super-Kamiokande Collaboration, Phys.Rev. D73 112001 2006
- [6] The SNO Collaboration, Phys.Rev. C72 055502 2005
- [7] S. Dazeley, A. Bernstein, N Bowden and R. Svoboda, To be published in Nucl. Instr. and Meth. A.
- [8] W. Coleman, A. Bernstein, S. Dazeley and R. Svoboda, Nucl. Instr. and Meth. A, vol. 595 pp. 339-345, 2008
- [9] X. Dai, E. Rollin, A. Bellerive, C. Hargrove, D. Sinclair, C. Mifflin, F. Zhang, Nucl. Instr. and Meth. A, vol. 589 pp. 290-295, 2008